

Lawrence Berkeley National Laboratory

Recent Work

Title

SINGLE AND DOUBLE ELECTRON CAPTURE BY 7.7 - TO 166-keV³ He⁺⁺ IONS IN Ng

Permalink

<https://escholarship.org/uc/item/5qt192zm>

Author

Steams, John Warren.

Publication Date

1967-09-11

C. 2

University of California Ernest O. Lawrence Radiation Laboratory

SINGLE AND DOUBLE ELECTRON CAPTURE
BY 7.7- TO 166-keV $^3\text{He}^{++}$ IONS IN N_2

John Warren Stearns
(M. S. Thesis)

September 11, 1967

RECEIVED
LAWRENCE
RADIATION LABORATORY
SEP 11 1967
LIBRARY AND
DOCUMENTS SECTION

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 5545*

Berkeley, California

UCRL-17756
eq. 7

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

UNIVERSITY OF CALIFORNIA
Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

SINGLE AND DOUBLE ELECTRON CAPTURE
BY 7.7- TO 166-keV ${}^3\text{He}^{++}$ IONS IN N_2

John Warren Stearns
(M. S. Thesis)

September 11, 1967

Contents

Abstract	ii
I. Introduction	1
II. Theory	5
A. Classical Capture	7
1. Thomas	7
2. Gryzinski and Others	7
B. Quantum Mechanical Capture	8
C. Double Capture	10
1. Classical	10
2. Quantum-Mechanical	10
III. Experiment	
A. General	11
B. Apparatus and Method	11
C. Data Reduction	11
1. Equations and Approximations Used for Data Reduction	15
2. Experimental Considerations in Data Reduction	19
3. Final Estimates	20
IV. Results and Discussion	23
A. Results of This Experiment	23
B. Comparison With Other He ⁺⁺ Experiments	23
C. Comparison With Theory	26
D. Suggestions for Future Experiments	26
Acknowledgments	29
Appendix	30
References	32

SINGLE AND DOUBLE ELECTRON CAPTURE
BY 7.7- TO 166-keV ${}^3\text{He}^{++}$ IONS IN N_2

John Warren Stearns

Lawrence Radiation Laboratory
University of California
Berkeley, California

September 11, 1967

ABSTRACT

The cross sections, σ_{21} and σ_{20} , for single and double electron capture of ${}^3\text{He}^{++}$ ions with energies between 7.7 and 166 keV have been measured in thin targets of N_2 . The ${}^3\text{He}^{++}$ ions were extracted from an rf source and accelerated electrostatically. After momentum analysis by a 90-deg magnet, they passed through the target chamber and were reanalyzed electrostatically. The charged components, ${}^3\text{He}^{++}$ and ${}^3\text{He}^+$, were detected by a pair of Faraday cups, and the neutral component ${}^3\text{He}^0$ by a CsI(Tl) crystal mounted on a photomultiplier tube. Currents from these detectors were simultaneously integrated, and measurements were made for at least eight pressures at each energy. Pressures were measured with a capacitance manometer. Both σ_{21} and σ_{20} have maxima at about 50 keV, which are $\sigma_{21} = (13.0 \pm 1.3) \times 10^{-16}$ and $\sigma_{20} = (3.3 \pm 0.7) \times 10^{-16}$ $\text{cm}^2/\text{molecule}$. Our results agree with those of Allison for ${}^4\text{He}^{++}$ in the region where the velocities overlap (${}^3\text{He}^{++}$ energies between 112.5 and 166 keV). The results are compared with available theoretical models.

I. INTRODUCTION

The process of electron capture or of electron loss via collisions with atoms, ions, or molecules has been a subject of interest for a long time. Rutherford¹ performed some of the earliest experiments and Fowler² presented one of the first theories on the capture of electrons from neutral gas by ions.

Subsequently, Thomas,³ Oppenheimer,⁴ Brinkman and Kramers,⁵ Jackson and Schiff,⁶ and many others have studied the problem theoretically. A summary of theoretical and experimental aspects of electron capture may be found in a book edited by Bates.⁷ Most previous work has been done for capture by protons; however, attempts to understand more complicated systems are now in progress. The present work is concerned with single and double capture by He^{++} in N_2 for the tens of keV range.

Aside from interest to atomic physicists this subject is important to many disciplines. For example, proposed thermonuclear reactors will contain very hot plasmas to produce fusion reactions. One important problem in such a reactor is the loss of energetic ions via charge exchange with slow neutrals. The fast neutral atom thus produced can leave the magnetic field that normally confines the plasma. It then becomes a slow neutral upon striking the wall of the vacuum chamber, or sputters material from the wall that can enter the plasma to perpetuate the process.

The dynamics of the sun cannot be fully understood without knowledge of rates of ionization and recombination. The sun regularly spews protons and α particles into space. Those that reach the

vicinity of the earth are often found to be neutral hydrogen and helium atoms or He^+ . One question is how and where were these ions neutralized and what can be inferred about the sun and the space between. These ions, and neutrals which become reionized, produce various atmospheric phenomena such as aurora. Detailed knowledge of capture and loss probabilities is necessary to understand these.

A heavy-ion accelerator has a section called a "stripper," where preaccelerated ions are stripped of some of their remaining electrons through collisions with atoms in a vapor or a foil.⁸ The efficiency of this stripper is directly related to the various probabilities of capture and loss of electrons. A tandem Van de Graaff or "swindletron" takes advantage of charge exchange and ionization by accelerating negative ions from a grounded terminal to a positive high-voltage terminal, where they are doubly (or more highly) stripped to form positive ions which are then further accelerated toward another grounded terminal. The energy of the ions is greater than it would be if they were originally produced at the high-voltage terminal.

Finally, all accelerators have the problem of loss of beam by charge exchange of the beam ions with the background gas during acceleration. This is especially important at certain low energies which the ions must pass through during their acceleration.

The general process of capture of electrons can be expressed uniquely for each combination of ion, gas, and energy in terms of a "cross section." A cross section, as the name implies, has units of area per target atom or molecule.

Let $\sigma \equiv$ the cross section for a reaction,

$p \equiv$ the probability for that reaction in a vanishingly small length dl ,

and $n \equiv$ number per unit vol = number density of the target gas; then we have the relation

$$p = n\sigma dl,$$

which defines σ in terms of the measurable p . For the process of charge exchange, we use the symbol σ_{if} , which denotes the cross section for changing the beam ion (or atom, molecule, etc.) from the charge state i to the charge state f in a single encounter with one of the target atoms or molecules.

In an investigation of charge exchange involving a He beam, generally three charge states are involved: He^0 , He^+ , and He^{++} . He^- can also exist (in metastable states), but its probability of production by electron capture is so small that its presence can be neglected in experiments in which the other components are of primary interest.^{9,10} Normally, six cross sections are involved: σ_{20} , σ_{02} , σ_{21} , σ_{12} , σ_{10} , and σ_{01} . One can readily see that the many competing processes could blot out the measurement of any one of them, depending upon the relative values of these cross sections and on other factors such as purity of charge species in the beam or target "thickness." (Thickness is defined as the product of the density and the length of the target.) (Thickness $\equiv \pi = nl$.) Fortunately (see Sec. III C), this interference due to multiple cross sections becomes nil in the limit as $\pi \rightarrow 0$ for an incident beam of one species.

Several experimentalists have explored single and double capture by He^{++} ions in nitrogen or air, i.e., at energies above 150 keV (e.g., Refs. 1, 11, 12). Previous to the work presented here it was not known how to extrapolate the values for the cross sections below this energy. The experimental work presented here shows a peak in each cross section measured (σ_{21} and σ_{20}) at about 50 keV ^3He energy). The single capture cross section, σ_{21} , trails off fairly steeply at greater and lesser energies, while the double capture cross section, σ_{20} , is nearly level at low energies, rising only slowly to its peak, and then trailing off nearly parallel to the other cross section at higher energies.

II. THEORY

A theory predicting charge-exchange cross sections accurately for all energies and all gases and ions has long eluded physicists. Even the simple case of neutralizing a proton by charge exchange in atomic hydrogen has proven quite difficult theoretically. At low velocities, the complications that arise from having to consider the temporary molecular states of the target atom and colliding ion system make the problem formidable. However, even approximations for velocities higher than the orbital electron velocity differ from one another by factors of two. A further complication arises when one tries to compare theory with experiment, since an atomic hydrogen target is difficult to prepare, whereas a molecular target is difficult to treat theoretically. As a result people have been forced to use various approximations. A qualitative low-energy model is afforded us by Massey's adiabatic hypothesis,¹³ which treats slow encounters, and also predicts large cross sections at nearly zero energy for "resonance" reactions, those in which the total internal energy of the system remains unchanged.

At high energies the Born approximation, based on quantum mechanics, and "impulse" approximations (classical or quantum mechanical) have been used. These approximations require a sudden transition between the initial and final states, allowing for no transitory states between.

So far, only semi-empirical methods have been found to treat the intermediate energies.

A simple way of looking at capture qualitatively has been afforded us through a bit of logic called "Massey's Adiabatic Hypothesis." For a nonresonant reaction it is Massey's argument that at rather low energies the approach of an oncoming particle can be "felt" by the target particle in such a way that the electronic configuration can adjust itself more or less adiabatically to the changing forces so that capture becomes improbable. This is because the energy difference between an electron belonging to one atom or the other cannot be transferred to any of the particles involved in an adiabatic transition.

At higher energies, the encounter becomes less adiabatic and the cross section rises. At very high energies, another process comes into play. An encounter may be over so quickly that an electron does not have enough time to react to the changing potentials and to be captured. Hence one would expect that for rather high energies the capture cross section would fall off, which, indeed, at high energies it does.

Now with these two processes acting to reduce the cross section at low and at high energies, one would expect to find an energy for which the cross section is a maximum. This peak should correspond in some way to the energy which needs to be absorbed (or supplied) in the reaction, being at greater relative velocities for greater energy differences.

Massey has deduced a simple expression to predict this peak based on the energy difference between the initial and final states of the system at the moment of reaction. For an energy difference, ΔE , called by Massey the "energy defect" in the reaction $A^+ + B \rightarrow A + B^+$,

Massey predicts that for some collision parameter, ℓ , the peak in the cross section should occur at the velocity

$$v = \frac{\ell \Delta E}{h},$$

where h is Planck's constant. In units of eV this becomes

$$T = 36(\Delta E)^2 m \ell^2 \text{ eV},$$

for m in amu and ℓ in Bohr radii. It will be noted that this simple expression is really quite difficult to apply, because ℓ can only be guessed at. The manner in which it varies with the charge and configuration of the two particles is unknown. Moreover, ΔE , which is usually assumed to be the difference in binding energies of the separated systems, could change during the approach. From experience with the measurements of several types of cross sections, one might get a feeling for the range over which a peak might occur, but beyond that the expression is not of much help.¹⁴

A. Classical Capture

1. Thomas

More detailed pictures of single capture have evolved principally for capture at the higher energies, where adiabatic transitions do not occur. Thomas in 1927 proposed a model³ that involved a double collision in which the incoming particle first struck an electron, driving it toward the target nucleus, where it rebounded in such a way as to find itself bound to the incident particle.

2. Gryzinski and Others

Since 1959 Gryzinski¹⁵ and others have presented modifications on

the work of Thomas by adjusting the limits of integration or by assuming different electronic configurations. The results of the modified theories have more closely approached the measured values. Bates and Mapleton^{16,17} have continued with this type of approach and have achieved remarkable success for energies somewhat higher than ours. Mapleton¹⁸ has calculated cross sections for us to be compared with our measurements of σ_{21} . His results are shown in Fig. 3, along with others.

B. Quantum Mechanical Capture

In 1928 Oppenheimer⁴ calculated the capture cross section between Hydrogen and He^{++} ions, employing the "overlap" integral

$$\epsilon = \int d\tau \psi_{\text{H}} \psi_{\text{He}^{++}}^* \psi_{\text{H}^+}^* \psi_{\text{He}^+}^* .$$

Calculations involving the use of this type of integral usually are approximated by using spherical waves for the bound electron and plane waves for the oncoming ion and outgoing particle. With an interaction potential and the proper normalization, the expression can be made into an infinite series called the Born approximation, which can be used to evaluate a capture cross section. Most calculations employ only the first term of this approximation. There is some question whether, for capture, this series converges.

In 1930, Brinkman and Kramers did a more precise calculation based on the first Born approximation.⁵ They derived the expression

$$\sigma = a_0^2 \frac{2^{18} \pi}{5} Z^5 Z'^5 S^8 [S^2 + (Z + Z')^2]^{-5} [S^2 + (Z - Z')^2]^{-5} ,$$

where $X = v\hbar/2\pi e^2$, a_0 is the radius of the first Bohr orbit, Z and Z' are the effective charges of the target and the projectile respectively, v is the relative velocity, and \hbar is Planck's constant. This expression represents the cross section for capture from an s state into another s state, and does not take into consideration any interaction between nuclei.

Much debate has ensued among the theoreticians as to whether the interaction between the nuclei has any appreciable effect on the cross sections (see, e.g., Bohr,¹⁹ Jackson and Schiff,⁶ and Pradhan²⁰). So Jackson and Schiff published a paper calculating this effect. Their calculation showed that for capture from a hydrogen-like atom the cross section should be lower than previous calculations by a slowly varying factor,

$$\sigma_{J-S} = (0.37 - 0.66)\sigma_{B-K},$$

where the smaller value is appropriate at low energies (around 1 MeV or less) and the larger one applies only at very high energies. Bates and Dalgarno²¹ exploited this weak energy dependence and, using methods first developed by Saha and Basu,²² produced Brinkman and Kramers type of calculations for the capture cross section from the $1s$, $2s$, and $2p$ states into any state of the projectile. Although Jackson and Schiff give better results, it is generally agreed that the inclusion of the internuclear interaction is incorrect.

C. Double Capture

1. Classical

Most of the theoretical methods mentioned so far deal only with the capture of a single electron from the target particle. The capture of two electrons in a single encounter is difficult to treat and, except for the general considerations that should be valid for all capture as expressed in the adiabatic hypothesis, no "classical"-type calculations have been made.

2. Quantum-Mechanical

Several calculations involving quantum mechanical treatment of double capture have been made. Fulton and Mittleman²³ have calculated the reaction $\text{He}^{++} + \text{He}^0 \rightarrow \text{He}^0 + \text{He}^{++}$. Their calculations involve substituting the product of two one-particle exchange amplitudes for the two-particle exchange amplitude in a Born approximation.

Low-energy calculations have also been made for this same reaction.^{24,25} There is only one nonresonant calculation that would apply to He^{++} in N_2 . Evaluations of a theory by R. K. Janev were made for this case,²⁶ but they showed little relationship to the measured values, perhaps due to the large difference in binding energies of the first and second electrons in He.

III. EXPERIMENT

A. General

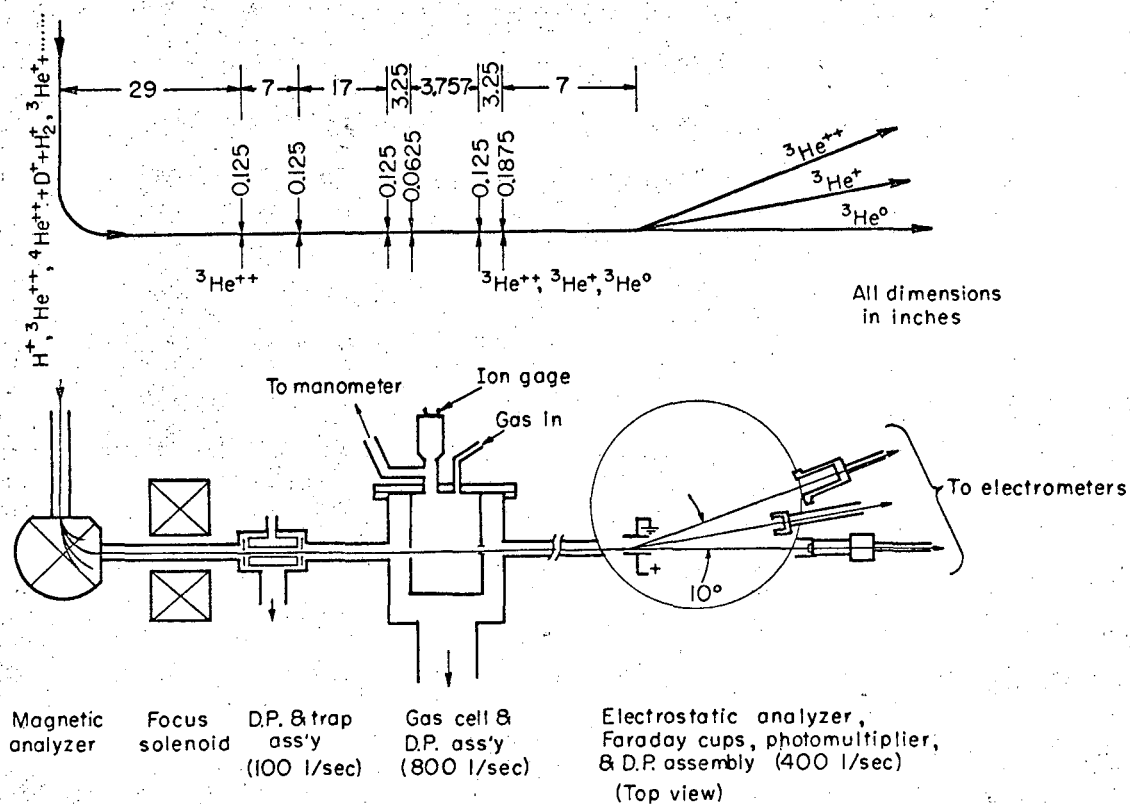
The experiment was performed by use of a modification of the "growth" method, in which the cross section is determined by observing the growth of the relative intensities of the product beams as gas is introduced into the path of the primary beam. This growth is approximately linear at low relative intensities, and is proportional to the pressure and the appropriate cross section. In order to extend these results significantly beyond the background contribution, a modified "growth" method was used.

B. Apparatus and Method

The experimental arrangement is shown in Fig. 1. He^{++} ions were produced in an rf ion source, which was placed at the top of an electrostatic acceleration column. The beam could be focused by an electrostatic "Einzel" lens, and the total accelerating voltage could be varied from about 3 kV to a maximum of about 110 kV.

The beam was analyzed by a 90-deg bending magnet and could be steered by a set of electrostatic deflectors. After being bent, the beam was refocused by a solenoid.

The beam then passed through a "gas cell" where the target was contained. The gas cell consisted of an inner chamber with small constrictions (63-mil input and 125-mil output diameters) for the beam, and a "differential" section which was being continuously pumped out. The nitrogen gas, used for the target, was admitted through a needle valve to maintain target pressures from 0.05 to 5 μ . The pressure was measured by a VG1A ion gauge and by a "Baratron" electromechanical



XBL676-3287

Fig. 1. Experimental arrangement. The upper diagram shows the positions and dimensions of the various collimators and constrictions along the beam path. The lower diagram shows the locations of the diffusion pumps and elements of the apparatus.

manometer. The two measurements agreed to within 5% in the overlapping range from 0.1 to 1.0 μ . The Baratron was also checked for calibration and linearity by an oil manometer at higher pressures. The pressure in the differential region was measured by a VGIA ion gauge and typically was less than one thousandth of the target pressure.

After passing through the target chamber, the beam was analyzed for reaction products by an electrostatic deflector. The two charged portions (He^{++} and He^+) were deflected into two Faraday cups and the neutral part impinged upon a CsI(Tl) crystal mounted on a photomultiplier tube. The secondary electron emission from the Faraday cups was suppressed by a small transverse magnetic field.

An electron current proportional to the neutral beam was produced by the photomultiplier. This current and the two produced at the Faraday cups were detected and amplified by three Keithly 410 electrometers. Signals from the electrometers were then used to drive three electronic integrating circuits.

The deflector could be turned off, causing the directly measurable charged portion of the beam to strike the neutral detector. The added current coming from the photomultiplier was then used to calibrate the detector.

The scintillator had a tendency to change sensitivity with time due to radiation damage, so it was necessary to calibrate it after each data measurement. These calibrations of the neutral detector showed large variations ($\pm 15\%$) as well as a time dependency, perhaps due to fluctuations in the beam. Thus the percent of uncertainty

associated with a single measurement of the neutral component of the beam was much greater than that for either of the other components or for the total beam (generally consistent to about 95% or better).

Due to contamination from having previously used the source with H_2 and D_2 , it was decided to do the experiment with ${}^3He^{++}$ to avoid confusion between ${}^4He^{++}$ and H_2^+ or D^+ . The electrometers could measure currents smaller than 10^{-13} A and with a 90% 4He -10% 3He mixture in the source the beam current was usually greater than 10^{-12} A.

For a cross-section measurement at any one energy at least 10 points were taken at pressures from background ($< 10^{-5}$ torr) to 1.5μ . The time for measuring each point was typically 20 to 30 sec.

The energy was considered to be twice that of the accelerating potential measured from the ion-source repeller. The voltage divider used for this purpose was calibrated to within 5% of the true voltage.

For each energy, the beam was first tuned to its maximum yield in ${}^3He^{++}$. The two charge integrators were checked for the correct relative integrating rates, the electrometers zeroed, and then several integrations made of the beam to find the background yield of He^0 and He^+ . Immediately after each data run (in which all three beams were integrated), a calibration run was made to measure the relative sensitivity of the neutral detector. This was done for every pressure by turning off the electrostatic deflector in the final analysis section and allowing the charged beams to strike the neutral detector. A timer was used which was gated on and off with the integrators to provide a basis for determining the amount of charged beam used for the calibration. This portion of each run also provided information on

the electronic drift of the integrators (always negligible, but accounted for in the analysis anyway). Occasionally, usually before and after changing to each new energy, the beam was removed from all detectors to determine the effect of dark current in the photomultiplier.

Before any gas was let into the gas cell, the Baratron manometer was zeroed. Then the gas was admitted and the pressure allowed to come to equilibrium. One or two measurements were made at each pressure, and after the highest pressure measurement was completed, a measurement or two was again made at lower pressures and at background pressure as a check.

The data were reduced by first calculating the percentage of each species with respect to the total beam. These points were then plotted against the pressure in the cell to give a preliminary determination of the cross sections (Fig. 2). These partially reduced data were then analyzed according to the method described in the next section.

C. Data Reduction

1. Equations and Approximations Used for Data Reduction

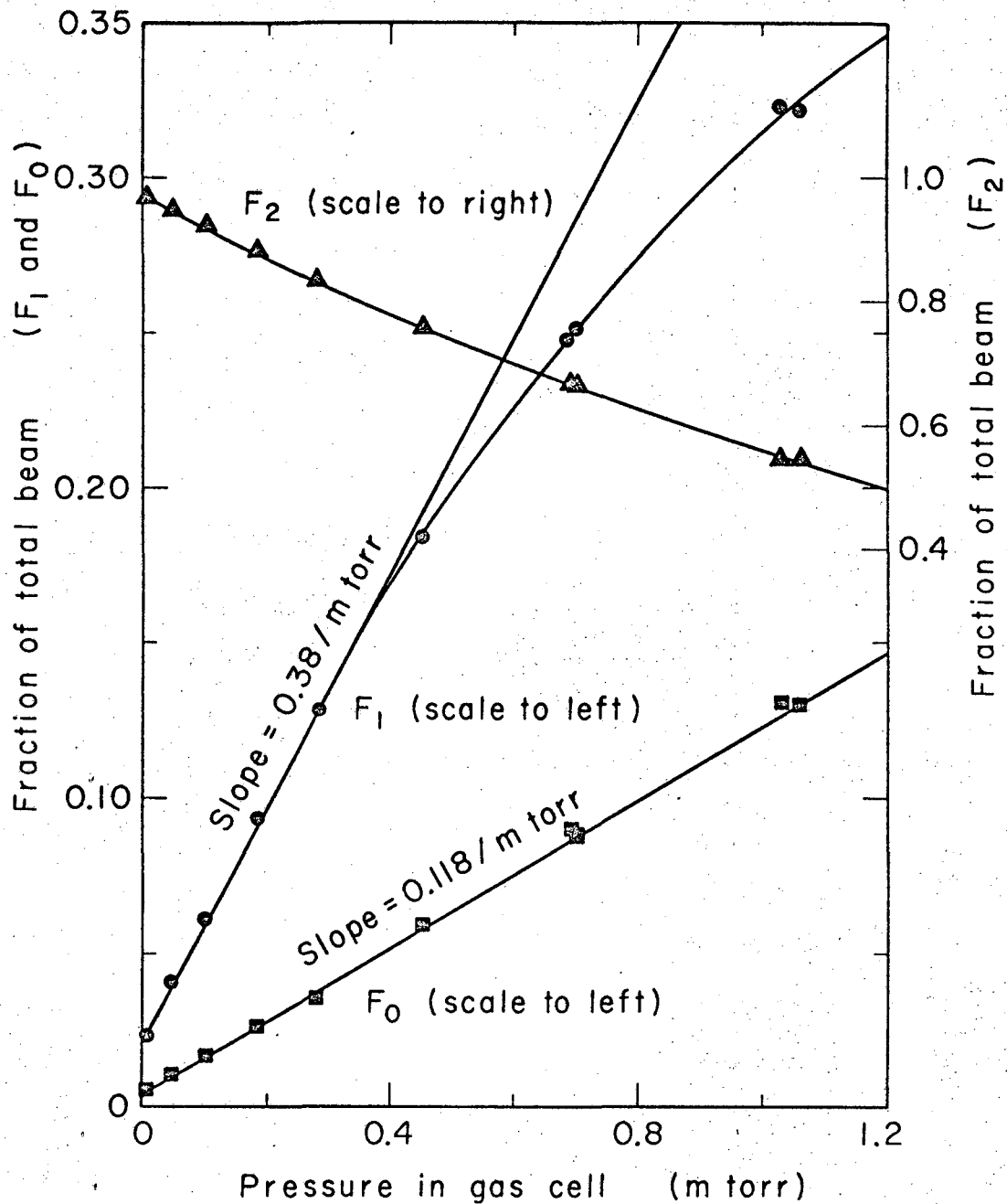
Let σ_{if} = the cross section per target particle (in cm^2) for the reaction that changes a beam particle from charge state i to charge state f ,

π = target "thickness" = n_l = number density x length of target,

F_i = measured fraction of beam of charge state i .

Assume: Beam entering target is entirely of charge state 2 (and fraction = 1).

Then we have the equations



XBL679-3819

Fig. 2. Plots of the uncorrected fractions of the beam vs pressure at 42 keV. The data in this example were used to calculate the points shown in Fig. 3. $F_2 = I_{(2+)}/I_{tot}$, $F_1 = I_{(+)}/I_{tot}$, and $F_0 = I_{(0)}/I_{tot}$.

$$dF_0/d\pi = -F_0(\sigma_{01} + \sigma_{02}) + F_1\sigma_{10} + F_2\sigma_{20}, \quad (1a)$$

$$dF_1/d\pi = F_0\sigma_{01} - F_1(\sigma_{10} + \sigma_{12}) + F_2\sigma_{21}, \quad (1b)$$

$$F_0 + F_1 + F_2 = 1. \quad (1c)$$

The complete solution was worked out by Allison.⁹ The solution is

$$F_i = F_{i\infty} + P(z, i)e^{\pi q} + N(z, i)e^{-\pi q} \exp \left[-\frac{1}{2} \pi \sum \sigma_{if} \right], \quad (2)$$

where $F_{0\infty} = (f\sigma_{21} - a\sigma_{20})/(ag - bf)$

and $F_{1\infty} = (b\sigma_{20} - g\sigma_{21})/(ag - bf),$

$$F_{2\infty} = \frac{[\sigma_{20}(a - b) + g(a + \sigma_{21}) - f(b + \sigma_{21})]}{(ag - bf)},$$

$$a \equiv -(\sigma_{10} + \sigma_{12} + \sigma_{21}),$$

$$b \equiv (\sigma_{01} - \sigma_{21}),$$

$$f \equiv (\sigma_{10} - \sigma_{20}),$$

$$g \equiv -(\sigma_{01} + \sigma_{02} + \sigma_{20}),$$

$$q \equiv \frac{1}{2} [(g - a)^2 + 4bf]^{1/2},$$

$$\sigma_{if} = (a + g),$$

and for

$$F_1, P_{z1} = P_{21} = \frac{1}{2q} [F_{1\infty}(s - q) - bF_{0\infty}],$$

$$N_{z1} = N_{21} = P_{21} - F_{2\infty},$$

$$F_0, P_{z1} = P_{20} = P_{21}(s + q)/b,$$

$$N_{z1} = N_{20} = N_{21}(s - q)/b,$$

where $s \equiv \frac{1}{2}(g - a),$

This has been reduced to the linear and squared terms of the Taylor expansion by Fogel and Mitin,²⁷

$$F_0/F_2 = \pi\sigma_{20} + \frac{1}{2}\pi^2(\sigma_{21}\sigma_{10} + \sigma_{20}\sigma_{21} + \sigma_{20}^2 - \sigma_{20}\sigma_{01} - \sigma_{20}\sigma_{02}); \quad (3a)$$

by symmetry,

$$F_1/F_2 = \pi\sigma_{21} + \frac{1}{2}\pi^2(\sigma_{20}\sigma_{01} + \sigma_{21}\sigma_{20} + \sigma_{21}^2 - \sigma_{21}\sigma_{10} - \sigma_{21}\sigma_{12}). \quad (3b)$$

The known σ_{02} and σ_{12} are small so that they may be neglected:⁷

$$\sigma_{02} + \sigma_{12} \ll \sigma_{21} \text{ or } \sigma_{20}$$

(for all energies < 300 keV). For π small enough, $F_0/F_2 \approx \pi\sigma_{20}$ and $F_1/F_2 \approx \pi\sigma_{21}$. If we substitute these into the squared terms for F_0/F_2 we get

$$\frac{F_0}{F_2} = \pi\sigma_{20} + \frac{1}{2}\pi\frac{F_1}{F_2}\sigma_{10} + \frac{1}{2}\pi\frac{F_1}{F_2}\sigma_{20} + \frac{1}{2}\pi\frac{F_0}{F_2}\sigma_{20} - \frac{1}{2}\pi\frac{F_0}{F_2}\sigma_{01}.$$

Rearranging,

$$\pi\sigma_{20} = \frac{F_0 - (F_1/2)\pi\sigma_{10} + (F_0/2)\pi\sigma_{01}}{F_2 + \frac{1}{2}(F_1 + F_0)},$$

but from Eq. (1c) we get

$$F_2 = 1 - (F_1 + F_0),$$

hence

$$\pi\sigma_{20} = \frac{F_0 - (F_1/2)\pi\sigma_{10} + (F_0/2)\pi\sigma_{01}}{1 - \frac{1}{2}(F_1 + F_0)} \quad (4a)$$

Similarly

$$\pi\sigma_{21} = \frac{F_1 - (F_0/2)\pi\sigma_{01} + (F_1/2)\pi\sigma_{10}}{1 - \frac{1}{2}(F_1 + F_0)} \quad (4b)$$

Since σ_{10} and σ_{01} are known throughout the range of the experiment²⁸ (see Table I), we now have expressions in terms of known or measurable quantities which should be linear in π within the range of our approximation. The error associated with this approximation should be at most 4.2% for a beam attenuation of 39% or less. See the Appendix for an analysis of this error.

2. Experimental Considerations in Data Reduction

In the experiment, before Eqs. (4a and b) could be applied, certain considerations had to be made.

When the gas cell was entirely pumped out ($< 10^{-5}$ torr), a small background of He^+ and He^0 remained in the beam ($F_0 + F_1 \approx 2\%$ to 4%). If these were ignored, the plots of F_1 and F_0 , corrected according to the second-order effects in σ_{01} and σ_{10} , could have an incorrect slope, depending upon whether the backgrounds were produced ahead of the gas cell or not.

Actually, ignoring the background has the effect of assuming that one-half of the background originates before the target. Since the distinction is small, this seems to be a satisfactory compromise.

Hence to reduce the data it was necessary only to calculate the

fractions, plot $\pi\sigma_{21}$ and σ_{20} according to Eqs. (4a and b), and then find the slope of the best straight line through the points on the plot. This line was found with a least-squares fit routine (see Fig. 3).

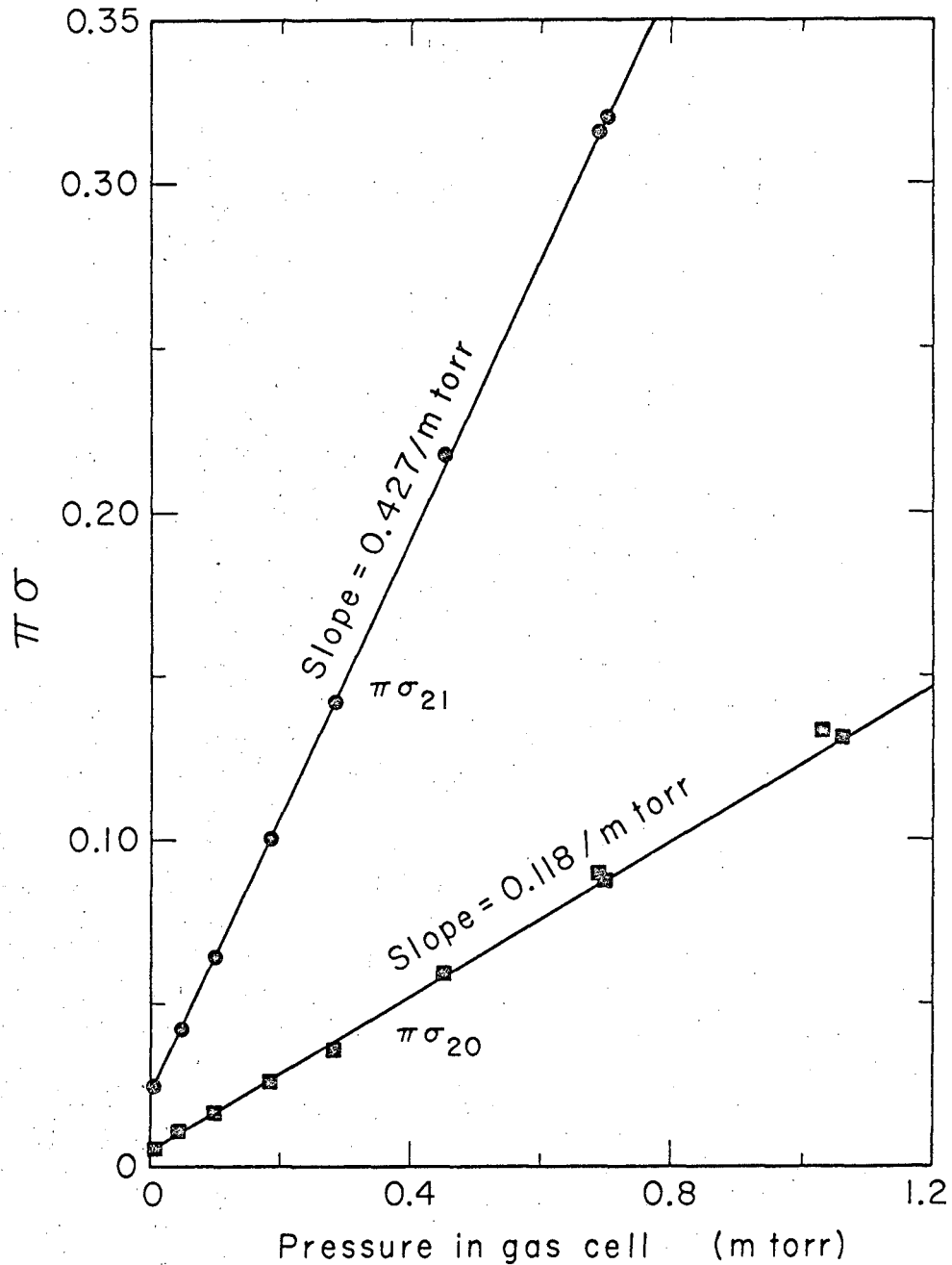
In order to check that the background actually had very little effect, calculations were made considering the background to have originated before the gas cell for one case and after the gas cell for another. These calculations were made at two different energies for which the background was relatively high. The two cases bracketed the uncorrected result and differed from it by less than 5% in σ_{20} , less than 1% in σ_{21} .

The effect of metastable states in He on the cross section σ_{01} has been discussed by Wittkower et al.²⁹ In order to see how an incorrect value for this cross section would affect the linearizing corrections, calculations for the worst possible case (a combination of high pressure in the gas cell and large σ_{01}) were made by using a value twice as great for σ_{01} . This appeared to affect the results by at most 9% for σ_{20} , and proportionately less for σ_{21} (1.5%).

3. Final Estimates

The possible error in data reduction is discussed in the appendix. The uncertainty associated with the electronics is about 3%. The greatest random error is associated with the neutral measurement, as previously discussed. This error is about 15% for each data point (ten or more data points per cross-section result given in Table I).^{*} This analysis gives a smaller uncertainty than seems apparent in the

^{*}The total random error is therefore about 5%



XBL679-3818

Fig. 3. Plots of $\pi\sigma_{21}$ and $\pi\sigma_{20}$ at 42 keV calculated from the data in Fig. 2, using Eqs. (4a) and (4b), Sec. III C. The points shown above 1 mtorr were not used in the least-square fit to determine the slope. Note that for this example, the slope of $\pi\sigma_{20}$ is the same as that of F_0 in Fig. 2, whereas the slope of $\pi\sigma_{21}$ is greater than that of F_1 in Fig. 2.

data (Fig. 4), hence it would seem prudent to assign about 10% error to the σ_{21} cross section and about 20% to the σ_{20} cross section. The source of this error is not known. The most significant systematic error is probably associated with the pressure measurements, which should probably not be relied upon more than to 7 or 8%. The estimated error in the energy is about 5%.

IV. RESULTS AND DISCUSSION

A. Results of This Experiment

The results are shown in Table I and Fig. 4. Both cross sections show a maximum at about 50 keV ^3He energy. The single-capture cross section (σ_{21}) has a maximum of about $13 \times 10^{-16} \text{ cm}^2$ and falls off fairly rapidly at higher and lower energies. The low-energy behavior of the double-capture cross section (σ_{20}) is somewhat indeterminate, but could be construed as starting to level off. At higher energies it rises slowly to a maximum of about $3.3 \times 10^{-16} \text{ cm}^2$ and then falls off at about the same rate as the single-capture cross section.

B. Comparison With Other He^{++} Experiments

Plotted with the results in Fig. 4 are those of Pivovar et al.,¹² Allison,⁹ Nikolaev et al.,¹¹ and Rutherford.¹ These results agree well in the overlapping region and appear to blend smoothly into the present results.

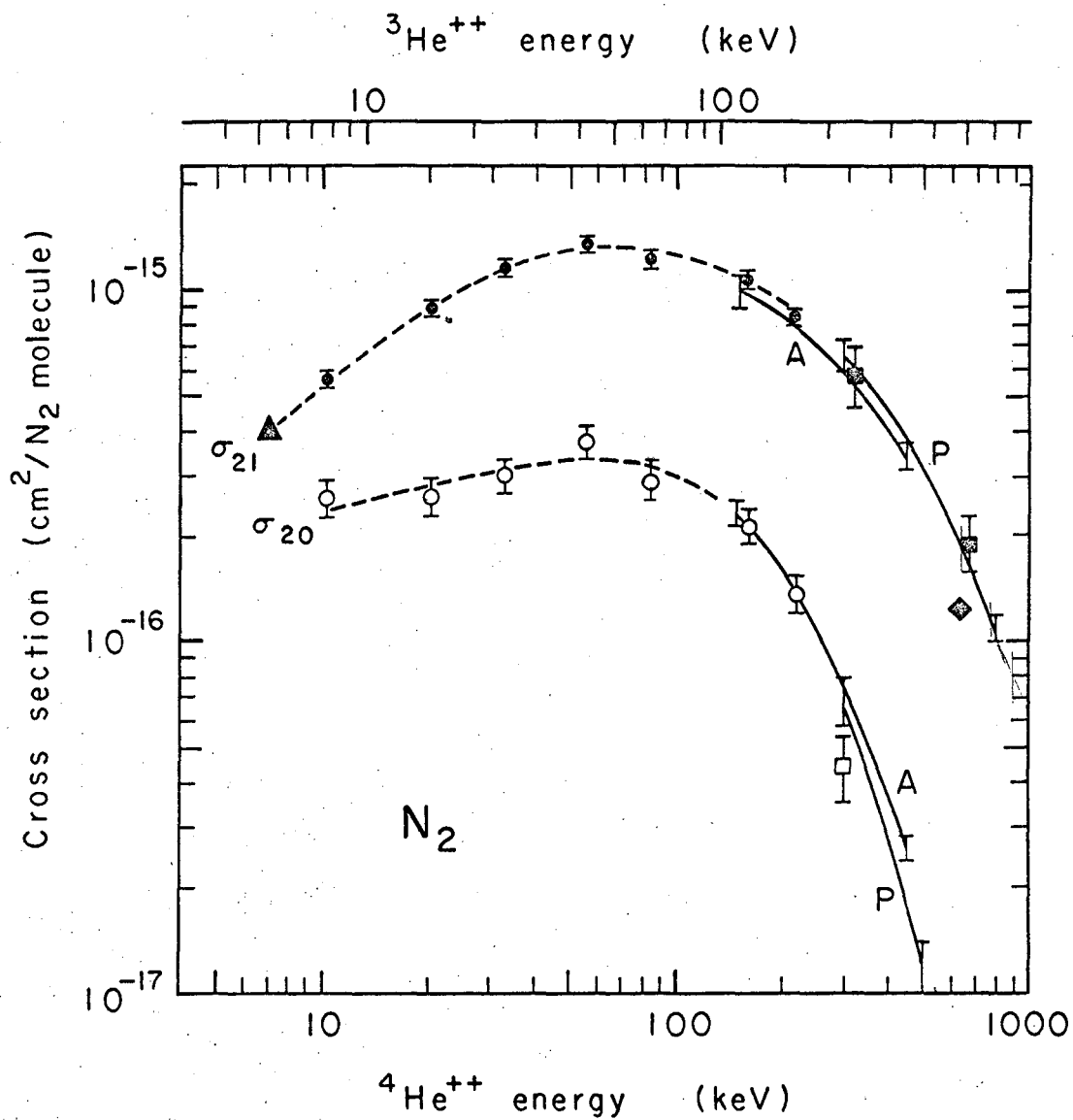
The results of Allison (solid line marked A) were deduced from equilibrium fractions and relative attenuations of the various species as gas was introduced into a region containing a magnetic field that was also used to analyze the equilibrium fractions. His measurements were done in air, which should yield very nearly the same results as pure nitrogen. The effect of metastables on his results should be negligible, since all the cross sections involved were deduced from the same set of data. The result of Rutherford (\diamond) is also for air.

Pivovar et al. (solid line P) used a method similar to ours in several gases. The results for N_2 are shown for comparison. Nikolaev et al. (\square , \blacksquare) counted particles in a method also similar to ours for

Table I. Measured values and uncertainties for σ_{21} and σ_{20} .

Actual energy (keV)	Energy per nucleon (keV/A)	σ_{21} (10^{-16} cm ²)	σ_{20} (10^{-16} cm ²)	σ_{10} (10^{-16} cm ²)	σ_{01} (10^{-16} cm ²)	Ion
7.2	1.8	$4.07 \pm 12\%$	< 4.0	4.1	0.14	${}^4\text{He}^{++}$
7.7	2.57	$5.55 \pm 10\%$	$2.61 \pm 20\%$	5.1	0.24	${}^3\text{He}^{++}$
15.3	5.1	$9.19 \pm 10\%$	$2.61 \pm 20\%$	6.5	0.64	${}^3\text{He}^{++}$
24.2	8.07	$11.6 \pm 10\%$	$2.98 \pm 20\%$	6.7	1.15	${}^3\text{He}^{++}$
42.0	14.0	$13.6 \pm 10\%$	$3.75 \pm 20\%$	6.2	2.05	${}^3\text{He}^{++}$
63.6	21.2	$12.21 \pm 10\%$	$3.10 \pm 20\%$	5.2	2.9	${}^3\text{He}^{++}$
117	39.2	$10.9 \pm 10\%$	$2.14 \pm 20\%$	3.4	3.3	${}^3\text{He}^{++}$
166	55.3	$8.3 \pm 10\%$	$1.39 \pm 20\%$	2.3	5.0	${}^3\text{He}^{++}$

The values for σ_{20} measured with 7.2 keV ${}^4\text{He}^{++}$ ions is only an upper limit due to an unknown amount of contamination from hydrogen isotopes; the value for σ_{21} was not greatly affected by the contamination. The values for σ_{10} and σ_{01} are from the work of Barnett and Stier,²⁸ and are estimated to be accurate to $\pm 10\%$. The uncertainty in all energies is $\pm 5\%$.



XBL678-3748-A

Fig. 4. Results of cross-section measurements for capture of one (σ_{21}) and two (σ_{20}) electrons by helium nuclei in N_2 . \blacktriangle , \bullet , \circ this paper; \blacklozenge Rutherford for air (Ref. 1); \square Nikolaev et al. (Ref. 11b); \blacksquare Nikolaev et al. (Ref. 11a); the line marked A presents the results of Allison for air (Ref. 9); the line marked P, Pivovar et al. (Ref. 12).

many multiply charged ions. His results for He^{++} in N_2 are shown.

C. Comparison With Theory

Figure 5 shows the results of evaluating the theories of Brinkman and Kramers,⁵ and an extension by Bates and Dalgarno.²¹ Also shown is the plot of a curve based on the classical formula of Bates and Mapleton,¹⁶ which was evaluated for us by Mapleton.¹⁸ The experimental points are shown for comparison.

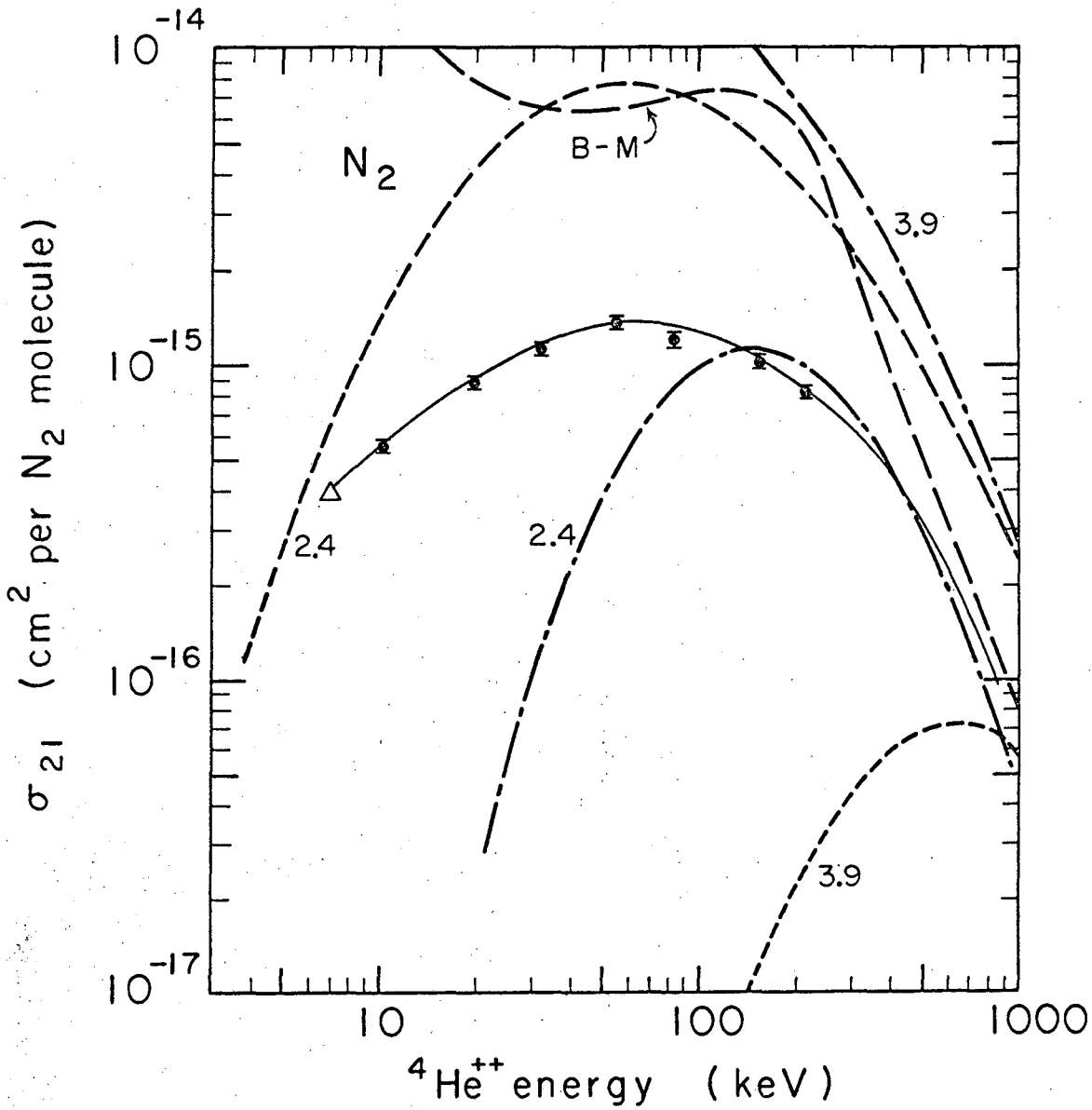
The Bates and Mapleton cross section includes capture from all states, whereas those of Brinkman and Kramers and Bates and Dalgarno are only for capture from the 2s and 2p states respectively.

All the theoretical results were based on atomic nitrogen and then doubled for comparison with the experiment.

Brinkman and Kramers suggested that for capture from nitrogen the effective Z should be 2 or 2.4. However, Slater³⁰ has suggested a prescription that would give an effective value of 3.9 for Z in this case. Curves based on $Z_{\text{eff}} = 2.4$ and 3.9 are shown for comparison. Also these values are used in the evaluations for 2p electron capture based on the Brinkman and Kramers type of capture of 2p electrons, which was presented by Bates and Dalgarno.²¹ Note that the low-energy predictions of the Brinkman and Kramers formulations are very strongly dependent on the choice for Z_{eff} .

D. Suggestions for Future Experiments

A look at the results shows the possibility of interesting structure in σ_{20} at the lower energies. Because of the uncertainties one cannot determine the low-energy trend. It would be interesting to repeat the last point or two and then to extend the measurements to



XBL676-4107-C

Fig. 5. Comparison of theory and experiment for σ_{21} in N_2 . The solid line summarizes the various experimental results shown in Fig. 4, and the points are the results of this paper. The theoretical predictions are: --- the Bates and Mapleton classical model (Ref. 12); --- the Brinkman-Kramers model for capture of the 2s electrons of nitrogen into the 1s state of He^+ (Ref. 5); and - · - the Brinkman-Kramers model for capture of 2p electrons into the 1s state of He^+ (Ref. 21). The Brinkman-Kramers curves are labeled with the values of Z_{eff} used in the calculation.

lower energies. Because of the many combinations of initial and final states, the adiabatic hypothesis would predict many minor peaks and perhaps some other major one at or near an accidental resonance.

Other gases would also be interesting, particularly He, since it would represent a resonant case for double capture. Further measurements need to be made also to determine the relative metastable populations for various cases and their relative cross sections.

Finally, other multiple capture cross sections might be considered for different ions of the same or greater charge. Particularly, other resonant and near-resonant cases might be interesting.

ACKNOWLEDGMENTS

I am very grateful to Dr. Robert V. Pyle for his continuing interest, guidance, and moral support, and to Dr. C. M. Van Atta for his support of this research. I am also thankful to Dr. Klaus H. Berkner for his participation in the experiment and for our many discussions, to Dr. R. A. Mapleton for providing us with numerical solutions for his classical model, and to Dr. M. H. Mittleman for his helpful discussions regarding theory. Many thanks also to Dr. Selig N. Kaplan for his earlier aid, to Vincent J. Honey for his aid with the apparatus, to Harlan A. Hughes, Louis A. Biagi, and their shop personnel for their help in constructing the equipment, and especially to Margaret R. Thomas for typing the manuscript.

APPENDIX

We will now estimate the errors associated with using only up to the second-order term to derive Eqs. (4a and b). Let $\sigma = \sigma_{21} + \sigma_{20}$, then the exact expression for F_2 is (still neglecting σ_{12} and σ_{02})

$$F_2 = e^{-\pi\sigma} = 1 - \pi\sigma + \frac{(\pi\sigma)^2}{2} - \frac{(\pi\sigma)^3}{2} + \frac{(\pi\sigma)^4}{2} - \dots, \quad (\text{A-1})$$

and the approximation error in $F_0 + F_1$ (expressed as a fraction), for the approximations (4a and b) is

$$\epsilon' < \frac{(\pi\sigma)^2}{6\left(\pi\sigma - \frac{(\pi\sigma)^2}{2}\right)} = \frac{(\pi\sigma)^2}{3(2 - \pi\sigma)}. \quad (\text{A-2})$$

Let $\pi\sigma = 0.5$, say (equivalent to a beam attenuation $> 39\%$),

$$\text{then } \epsilon' < \frac{0.25}{4.5} = \frac{0.5}{9} = 0.056. \quad (\text{A-3})$$

If we now approximate the error associated with substituting F_1 for $\pi\sigma_{21}$ we first note, for the same attenuation,

$$1 - F_2 = F_1 + F_0 \approx 0.39.$$

If we substitute $F_1 + F_0$ for $a(\pi\sigma)$ in the squared term--i.e., let $(\pi\sigma)^2 = (F_1 + F_0)\pi\sigma$ --we get, for our actual error,

$$\epsilon'' = \left| \frac{0.39 - [0.5 - (0.39 \times 0.5)/2]}{0.39} \right| < \frac{0.01}{0.39},$$

$$\epsilon'' = < 0.026 \rightarrow 2.6\%. \quad (\text{A-4})$$

Thus, except for the terms involving σ_{10} and σ_{01} , we expect an approximation error not greater than 2.6% for attenuations less than about 40%.

We will now estimate the error associated with linearizing the terms involving σ_{10} and σ_{01} . σ_{01} reaches a maximum of $6.7 \times 10^{-16} \text{ cm}^2$ at the point where $\sigma_{21} = 1.1 \times 10^{-15} \text{ cm}^2$ and $\sigma_{20} = 3.7 \times 10^{-16} \text{ cm}^2$. Thus for $\pi\sigma = 0.50$, $\pi\sigma_{21} = 0.33$ and $\pi\sigma_{10} = 0.25$.

If we assume that $\pi\sigma_{10}$ is the first term in the expansion

$$1 - e^{-\pi\sigma_{10}} = \pi\sigma_{10} - (\pi\sigma_{10})^2/2 + \dots,$$

then the maximum error associated with $\pi\sigma_{10}$ is

$$\epsilon'' = \left(\frac{F_1}{2} \frac{(\pi\sigma_{10})^2}{2} \right) / \pi\sigma_{21} < \frac{(\pi\sigma_{10})^2}{2}$$

Similar analysis for σ_{01} shows a smaller error for the same conditions (i.e., $\pi\sigma = 0.50$). Since σ_{01} and σ_{10} always work to oppose each other, the greatest possible approximation error then is

$$\epsilon < \epsilon'' + \epsilon''' = 2.6\% + 1.6\% = 4.2\%. \quad (\text{A-5})$$

REFERENCES

1. E. Rutherford, The Capture and Loss of Electrons by α Particles, Phil. Mag. 47, 277 (1924).
2. R. H. Fowler, Statistical Equilibrium with Special Reference to the Mechanism of Ionization by Electronic Impacts, Phil. Mag. 47, 257 (1924).
3. L. H. Thomas, On the Capture of Electrons by Swiftly Moving Electrified Particles, Proc. Roy. Soc. (London) A114, 561 (1927).
4. J. R. Oppenheimer, On the Quantum Theory of the Capture of Electrons, Phys. Rev. 31, 349 (1928).
5. H. C. Brinkman and H. A. Kramers, Zur Theorie der Einfangung von Elektronen durch α -Teilchen, Proc. Acad. Sci. Amsterdam 33, 973 (1930).
6. J. D. Jackson and H. Schiff, Electron Capture by Protons Passing through Hydrogen, Phys. Rev. 89, 359 (1953).
7. D. R. Bates, Ed., Atomic and Molecular Processes (Academic Press, New York, 1962).
8. R. L. Gluckstern, Electron Capture and Loss by Ions in Gases, Phys. Rev. 98, 1817 (1955).
9. S. K. Allison, Double Electron Capture and Loss by Helium Atoms Traversing Gases, Phys. Rev. 109, 76 (1958).
10. S. K. Allison, Electron Loss Cross Sections for Helium Atoms in the Kinetic Energy Range 100-450 keV, Phys. Rev. 110, 670 (1958).
11. V. S. Nikolaev, I. S. Dmitriev, L. N. Fateeva, and Ya. A. Teplova, (a) Experimental Investigation of Electron Capture by Multiply Charged Ions, Soviet Phys.-JETP 13, 695 (1961) [Zh. Eksperim. i

- Teor. Fiz. 40, 989 (1961)]; (b) Capture of Several Electrons by Fast Multicharged Ions, Soviet Phys.-JETP 14, 67 (1962) [Zh. Eksperim. i Teor. Fiz. 41, 89 (1961)].
12. L. I. Pivovarov, M. T. Novikov, and V. M. Tubaev, Electron Capture by Helium Ions in Various Gases in the 300-1500 keV Energy Range, Soviet Phys.-JETP 15, 1035 (1962) [Zh. Eksperim. i Teor. Fiz. 42, 1490 (1962)].
 13. H. S. W. Massey, Collisions between Atoms and Molecules at Ordinary Temperatures, Rept. Progr. Phys. 12, 261 (1949).
 14. For more discussions of the adiabatic hypothesis see, e.g., H. B. Gilbody and J. B. Hasted, Anomalies in the Adiabatic Interpretation of Charge-Transfer Collisions, Proc. Roy. Soc. (London) A238, 334 (1956).
 15. M. Gryzinski, Classical Theory of Electronic and Ionic Inelastic Collisions, Phys. Rev. 115, 374 (1959).
 16. D. R. Bates and R. A. Mapleton, Classical Calculations on Electron Capture, Proc. Phys. Soc. (London) 87, 657 (1966).
 17. D. R. Bates and R. A. Mapleton, On the Classical Theory of Electron Capture, Proc. Phys. Soc. (London) 90, 909 (1967).
 18. R. A. Mapleton (Air Force Cambridge Research Laboratories), private communication. These solutions are for the method described in Ref. 16.
 19. N. Bohr, The Penetration of Atomic Particles through Matter, Kgl. Danske Videnskab. Selskab Mat.-fys. Medd. 18, No. 8 (1948).
 20. See discussion in note 7 of T. Pradhan, Electron Capture by Protons Passing through Hydrogen, Phys. Rev. 105, 1250 (1957).

21. D. R. Bates and A. Dalgarno, Capture into Excited States in Encounters between Hydrogen Atoms and Fast Protons, Proc. Phys. Soc. (London) A65, 919 (1952).
22. M. N. Saha and D. Basu, Capture of Electrons by Positive Ions While Passing through Gases, Indian J. Phys. 19, 121 (1945).
23. M. J. Fulton and M. H. Mittleman, Double Charge Transfer, $\text{He}^{++} + \text{He}$, Phys. Rev. 138, A667 (1965).
24. D. Basu, S. C. Mukherjee, and N. C. Sil, Double Electron Capture of Alpha Particles Passing through Helium, in Atomic Collision Processes, M. R. C. McDowell, Ed. (North-Holland Publishing Co., Amsterdam, 1964), p. 769.
25. M. J. Fulton and M. H. Mittleman, Electron Capture by Helium Ions in Neutral Helium, Proc. Phys. Soc. (London) 87, 669 (1966).
26. R. K. Janev, Two-Electron Capture in Non-Resonant Heavy Particle Collisions, in Abstracts of Papers, V International Conference on the Physics of Electronic and Atomic Collisions, Leningrad, 1967 (Publishing House Nauka, Leningrad, 1967), p. 82; A Model of Double Non-Resonant Charge Exchange, Zh. Eksperim. i Teor. Fiz. 52, 1221 (1967).
27. Ya. M. Fogel and R. V. Mitin, Formation of Negative Hydrogen Ions by Collisions of Protons with Gas Molecules, Soviet Phys.-JETP 3, 334 (1956) [Zh. Eksperim. i Teor. Fiz. 30, 450 (1956)].
28. C. F. Barnett and R. M. Stier, Charge Exchange Cross Sections for Helium Ions in Gases, Phys. Rev. 109, 385 (1958).
29. A. B. Wittkower, G. Levy, and H. B. Gilbody, Metastable Atoms in Fast Helium Beams, Proc. Phys. Soc. (London) 90, 581 (1967).
30. J. C. Slater, Atomic Shielding Constants, Phys. Rev. 36, 57 (1930).

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

